DOI/ER-46023

Final Report

"X-ray Studies of Materials Dynamics at MHATT-CAT Sector 7 , Advanced Photon Source"

P.I.: Roy Clarke
University of Michigan, Ann Arbor, MI 48109-1040

U.S. Department of Energy Grant No.
DE-FG02-03ER46023

Project Period 12/01/02 to 11/30/05

SUMMARY

This Final Report describes the scientific accomplishments that have been achieved with support from grant DE-FG02-03ER46023 during the period 12/01/02 – 11/30/05. The funding supported a vigorous scientific program allowing the PI to achieve leadership in a number of important areas. In particular, research carried out during this period has opened way to ultrafast dynamics studies of materials by combining the capabilities of synchrotron radiation with those of ultrafast lasers. This enables the initiation of laser-induced excitations and studies of their subsequent dynamics using laser-pump/x-ray probe techniques. Examples of such excitations include phonons, shock waves, excitons, spin-waves, and polaritons. The breadth of phenomena that can now be studied in the time-domain is very broad, revealing new phenomena and mechanisms that are critical to many applications of materials.

Table of Contents

1. Overview of Research Mission at Sector 7, Advanced Photon Source

APS Sector 7, dedicated to time-resolved x-ray science, is operated by APS under a Partnership User (PU) agreement in which the UM receives a fraction of the beam time (10% during the project period) to develop longer term science programs and associated techniques in time-resolved x-ray science. The PI also has competed successfully for beam team time through the General User (GU) program at APS. In the past year the PI has been awarded 54 shifts (18 days) of general user time as well as sharing in the PU beam time.

The PI's group, through this partnership, is uniquely positioned to provide extensive training experiences for undergraduates, graduate students, and postdocs, as well as educational enrichment experiences for pre-college students. In particular, we have been successful in translating the excitement generated by the science we perform into a wider involvement of synchrotron users who are traditionally under-represented at national science facilities.

The PI's scientific program centers on real-time x-ray studies of materials and exploits the unique characteristics of the APS source. This research theme is made possible by the unprecedented brilliance of undulator radiation at the Advanced Photon Source, opening the door to new capabilities that were not previously possible:

- **studies of materials and devices under actual operating conditions**
- **exploiting the imaging capabilities of high-brightness x-ray beams**
- **using ultrafast lasers to coherently control states of matter and dynamics**
- **probing the microscopic behavior of interfaces and buried layers**
- **investigating length-scale dependent ordering and relaxation processes in real-time**

A wide range of experiments exploiting the special characteristics the APS have been successfully performed over the course of this award, some of which are in the "first of a kind" category. Some highlights include:

- First application of the Coherent Bragg Rod Analysis (COBRA) method for direct determination of interfacial atom arrangements in ferroelectric heterostructures (in collaboration with Hebrew University, University of Washington and Argonne National Labs Materials Science Division).
- Demonstration of x-ray refractive focusing optics using lithium.
- Time-resolved studies of phonon propagation and electron-hole plasma dynamics using ultrafast x-ray pump-probe techniques
- First studies of the dynamics of magnetorheological elastomers using speckle spectroscopy.
- First atomic-level structure determination of InAs-GaAs heterostructures using a direct, non-destructive method, COBRA.

This grant has supported research leading to 16 refereed journal publications, 11 refereed conference proceedings, and the Ph.D. dissertations of four graduate students.

2. Progress and Accomplishments under U.S. Department of Energy Award No. **DE-FG02-03ER46023**, December 1, 2002 – November 30, 2005.

Highlights of Research Accomplishments

2.1 Nano-rheology of MR elastomers

R. Clarke*[1], W. Schlotter*[1,2], Jevne Micheau*[1], John Ginder[3], M. Nichols[3], E. Dufresne[4].
[1]University of Michigan
[2]Stanford University
[3]Ford Research Lab
[4]Advanced Photon Source
*supported in part by this award

In collaboration with Ford Research Labs, we have studied the relaxation dynamics of a nano-magnetic composite material formed by embedding magnetic nanoparticles (ferrites) into a polymer (natural rubber). This class of materials, known as magneto-rheological (MR) elastomers [7] is of interest for their scale-dependent relaxation dynamics which can be controlled by an external magnetic field. There is a growing technological interest in applying MR materials as "smart materials" in a variety of applications ranging from magnetically controlled automotive suspensions and bushings, to active dampers for earthquake protection of tall buildings.
The magnetic nanoparticles in such a material form a linked, chain-like, network when the polymer matrix is cured in a magnetic field, as shown in Fig.1. Scanning electron microscopy (SEM) images of these networks provide insight into the static particle size distribution and chain structure. The response of the nanoparticle network to transient magnetic pulses is not presently well understood because of the difficulty of probing the motion of nanoparticles inside an optically opaque medium.

.

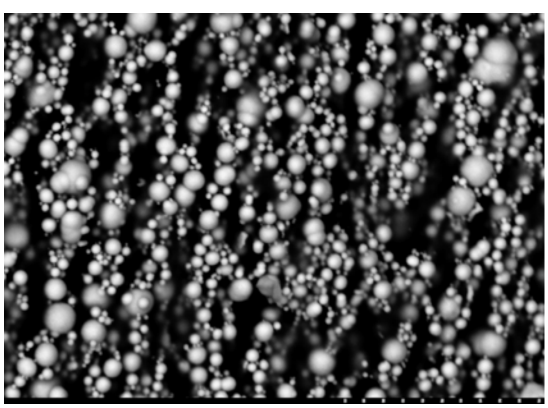


Fig.1: SEM image of magnetic particle network in a field-cured MR elastomer. Scale: 40x60 microns

For these experiments we have used ultrabright x-ray beams from the Sector 7 undulator to measure the small-angle transmission scattering of transversely coherent x-rays from the magnetic nanoparticles. Since the x-rays are transversely coherent (as in a laser) the phase of the wavefront will shift with the relative motion of the particles resulting in a time dependent x-ray interference pattern known as *speckle*. [8] By monitoring the time-dependent correlations of the x-ray speckle pattern we are able to determine the relaxation rate of the MR elastomer after the material is actuated by an external magnetic field. Since x-rays are able to probe density fluctuations down to the atomic scale this is an ideal probe for studying the length-scale dependence of relaxation rates; in other words to directly probe the magnetic contributions to the viscoelastic response. These first-of a kind experiments have revealed a wealth of information that was previously unobtainable on such materials. For example, we find that the relaxation rate decreases rapidly at length scales shorter than the particle-particle separation (~ 200nm) and is strongly field dependent. We have determined the scaling between the fluctuation of the x-ray correlations and mechanical properties such as shear modulus and viscoelasticity.   This topic formed the Ph.D. dissertation of Jevne Micheau-Cunningham, who graduated from the PI’s group in December 2004. Dr. Micheau-Cunningham joined Boston Scientific after his graduation.

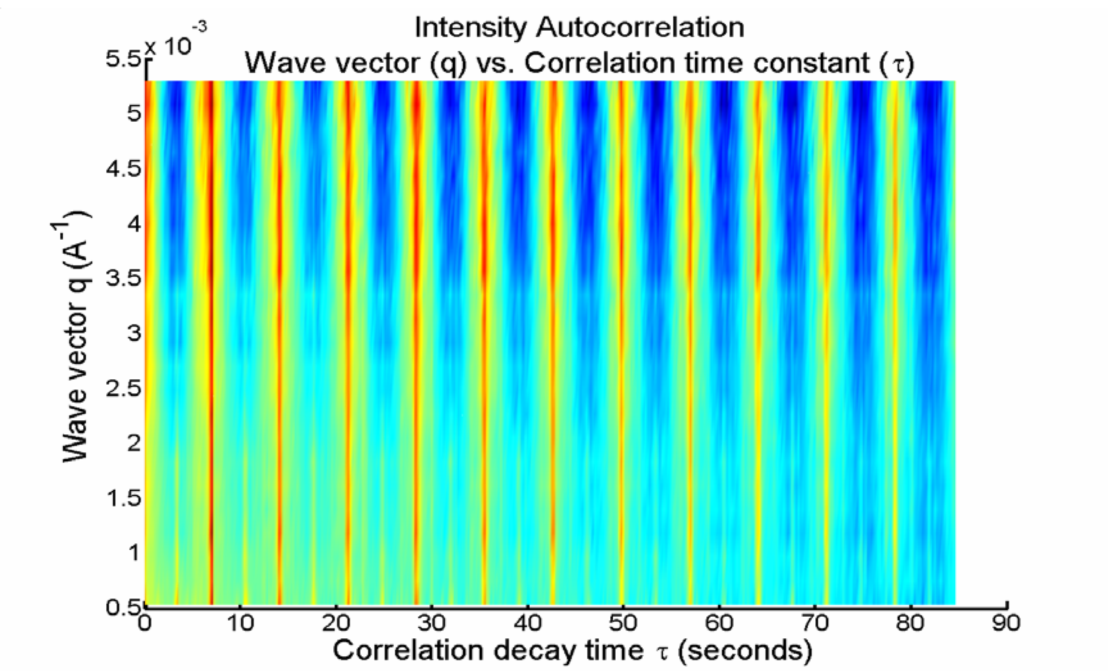


Fig. 2: Autocorrelation data for MR elastomer sample that is subjected to a pulsed magnetic field every 7 seconds. The strength of the autocorrelation is color coded with red as the strongest and blue as the weakest.   Note the long-time decorrelation of the particle motion which becomes more significant at higher wavevectors (smaller length scales). Note also the increase of the decay time (broadening of the vertical streaks), immediately after a pulse has been applied, as the dynamics is probed at larger wavevectors. This means that the relaxation rate is reduced at small length scales, which is the opposite of what is observed in Brownian motion.

The richness of the dynamic correlation data in Fig. 2 suggests novel approaches for the design of new magnetic nanoparticle composites whose viscoelastic properties can be tailored to suit a particular application by means of controlling the nanoparticle size distribution, and perhaps even in the future, the geometry of the nanoparticle networks.

2.2 Electron-hole plasma dynamics initiated by femtosecond laser pulses

M.F.DeCamp,[1] D.A.Reis,[1] A.Cavalieri,[1] P.H.Bucksbaum,[1] R.Clarke*,[1] R.Merlin,[1] E.M.Dufresne,[1] D. A.Arms,[1] A.M.Lindenberg,[2] A.G.MacPhee,[2] Z.Chang,[3] B.Lings,[4] J.S.Wark,[4] and S.Fahy [5]
*Supported in part by this award
[1]FOCUS Center and Department of Physics, University of Michigan
[2]Department of Physics, University of California, Berkeley
[3]Department of Physics, Kansas State University
[4]Department of Physics, Clarendon Laboratory, University of Oxford, Oxford, UK
[5]Physics Department and NMRC, University College Cork, Ireland.

We have studied coherent strain generated in (001)Ge by an ultrafast laser-initiated high density electron-hole plasma. The resultant coherent pulse is probed by time-resolved x-ray diffraction (see Fig. 3 showing the experimental arrangement at station 7-ID-D) through changes in the anomalous transmission of x-rays. The shock wave pulse front is driven by ambipolar diffusion of the electron-hole plasma and propagates into the crystal at supersonic speeds. Simulations of the strain including electron-phonon coupling, modified by carrier diffusion and Auger recombination, are in good agreement with the observed dynamics. This work has been published in Physical Review Letters.

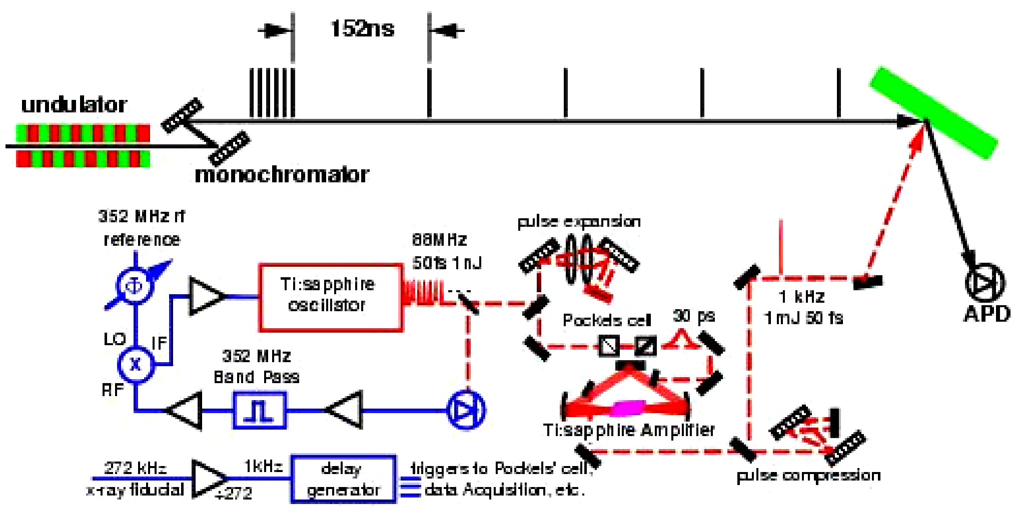


Fig. 3: Experimental setup for ultrafast diffraction measurements at Sector 7, APS, showing the single bunch pulse train incident on the sample and being detected by a gated avalanche photodiode detector (APD). The lower part of the diagram illustrates the laser/ x-ray synchronization and pump-probe delay (left); and laser amplifier (right).

The anomalous transmission of x-rays through the sample (essentially a time resolved version of the Borrmann effect is used as a probe of the strain front propagating through the crystal. The deflected-diffracted component of the x-ray beam emerging from the crystal (see Fig. 4) is modulated by the acoustic phonon excitation produced transiently by the femtosecond laser pulse incident on the back (emergent) side of the Ge crystal. One can observe a very fast (picosecond) transient which we associate with the production and subsequent diffusion of an electron-hole plasma. This fast transient behavior was monitored by placing an x-ray streak camera in the forward-diffracted beam at the exit position of the x-ray beam. This is shown in the inset of Fig. 4.

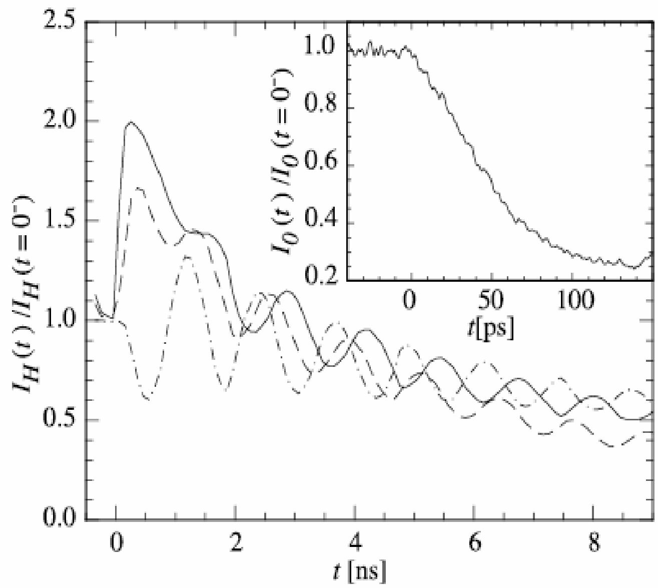


Fig. 4: Transient intensity changes in the deflected-diffracted x-ray beam, I_H, and forward diffracted beam I_O (inset, with a fluence of 35 mj/cm^2) as a function of laser pump-x-ray probe delay time, t. - - - is for low incident laser fluence of 2mJ/cm^2; _ _ _ corresponds to 7 mJ/cm^2, and the solid line 35 mJ/ cm^2.

The importance of this work is that it demonstrates, for the first time, that electron-phonon coupling can be used to study fast transient dynamics of electronic excitations through the strain that is induced in the lattice. The time-resolved x-ray probe used here is very sensitive to subtle changes in strain on picosecond time scales. Also of importance is the fact that current theoretical models (e.g., the Thomsen model) for energy transport in laser-matter interactions, do not adequately account for the short-time behavior that we observe. The results obtained here are useful input for improving the theoretical modeling of fast transient response of semiconductors to laser excitations. Developing applications of the time-dependent Borrmann effect will also be useful for studying the dynamics of other materials systems, such as ferroelectric thin film structures.

2.3 Ferroelectric Heterostructures

[In collaboration with Y. Yacoby (Hebrew University) and G.B. Stephenson (ANL).]

Graduate Students: Codrin Cionca and Vladimir Stoica;

Undergraduate Student: Jesse Guzman

Ferroelectric perovskites exhibit a number of unusual properties such as spontaneous electric dipole moments, large piezoelectric and pyroelectric coefficients, electrostriction and large nonlinear optical coefficients. Thin perovskite films in the form of epitaxial heterostructures are currently very interesting on account of the possibility that symmetry breaking at the heterostructure interfaces might influence the ferroelectric behavior. For example, when grown on a $SrTiO_3$ substrate (a para-electric material) $PbTiO_3$ (a ferroelectric) might exhibit additional symmetry breaking because of the presence of a biaxial strain due to coherent lattice matching at the interface between the film and the substrate. Also open questions remain as to the role of screening of the depolarization field by charges which accumulate at the film-substrate and film-environment interface.

My group has performed state-of-the-art x-ray measurements on this system at the Advanced Photon Source using a new structure mapping approach, COBRA (Coherent Bragg Rod Analysis). COBRA is a phase reconstruction technique utilizing two-dimensional x-ray scattering in the form of Bragg rods. It is capable of providing 3-dimensional maps of the thin film structure and the interface region with sub-Angstrom precision. Since the subtle atomic displacements responsible for ferroelectric symmetry breaking are at the level of fractions of an Angstrom this is a very promising approach for studying interfacial ferroelectricity. To our knowledge it is the only direct technique capable of providing these atomic scale details.

From theoretical considerations, behavioral modifications in the ferroelectric order parameter, polarization, are indeed expected near interfaces. In order to understand the properties of these films it is essential to know their structure in detail.

An example of our experimental results, recently published in Physical Review B, is shown in Fig. 5. On the $PbTiO_3$ side, the Ti atoms are displaced relative to the Pb atoms towards the $SrTiO_3$ substrate. Similarly, the O_2 and O_1 atoms are displaced

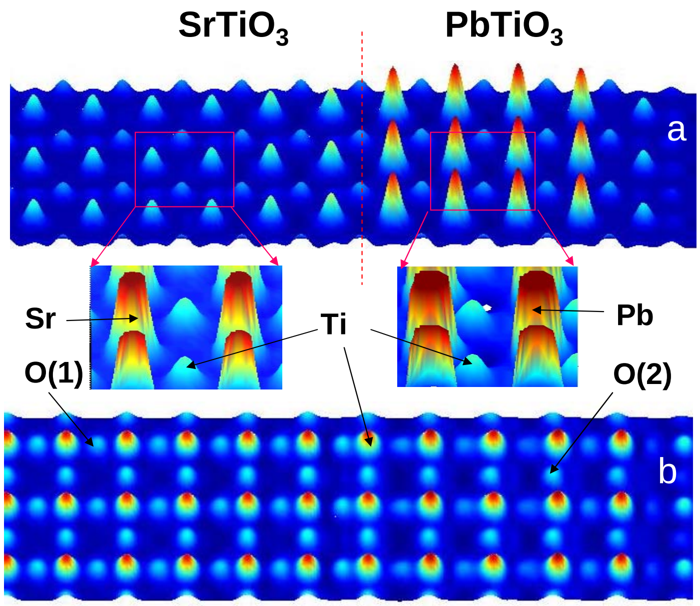


Fig. 5 COBRA map of PbTiO3 –SrTiO3 system. Shown here is (a) the plane perpendicular to the interface intersecting Sr, Pb, Ti, and O_1; (b) a plane perpendicular to the interface going through Ti, O_1 and O_2. The Ti-O_2 displacements can be seen clearly in the inset on the right.

Relative to the Ti atoms in the same direction. These results show that at ambient temperature (~450 K below the Curie point) the film is in a single domain ferroelectric state with the substrate providing a unique direction for the ferroelectric spontaneous polarization vector. These results clearly show that interface effects are responsible for the observed symmetry breaking and that epitaxial coherence can strongly affect the ferroelectric behavior.

Important details on the sublattice displacements that contribute to ferroelectricity in $PbTiO_3$ heterostructures can be extracted from COBRA maps similar to that shown in Fig. 5. For example, one can extract very precise quantitative values for the ferroelectric displacement of each sublattice. This is exactly the kind of information that is needed as input for first-principles calculation of the Born effective charges of each of the species in the structure. Our measurements indicate that the bulk values of the Born effective charges are not applicable in these ultrathin films, highlighting the need for such calculations. We are collaborating with theorists to address this question. Fig. 6 shows the integrated electron density and ionic displacements associated with each atomic species in the structure, including the two types of oxygen atoms O(1) and O(2). This is the first time that anyone was able to map the structure in such detail. This capability, enabled by the new COBRA phase construction technique supported under this grant, should be widely applicable to any epitaxial structure and represents an important step towards designing nanoscale ferroelectric devices in which the interfaces play such an important role.

Having now demonstrated that COBRA is indeed capable of revealing the subtle features that contribute to the ferroelectric behavior of these ultrathin films (only 4 monolayers thick), we are ready now to make COBRA measurements under dynamic conditions in the continuation of the project. We are particularly interested in finding out what is the behavior of each sublattice during switching of the electric polarization. Important questions relating to this are: first, how does the substrate contribute to establishing polarization in the ferroelectric film, especially the oxygen sublattices which seem to have the largest contribution; and second, what is the physics of switching fatigue in these materials such that after many switching cycles the magnitude of the polarization diminishes.

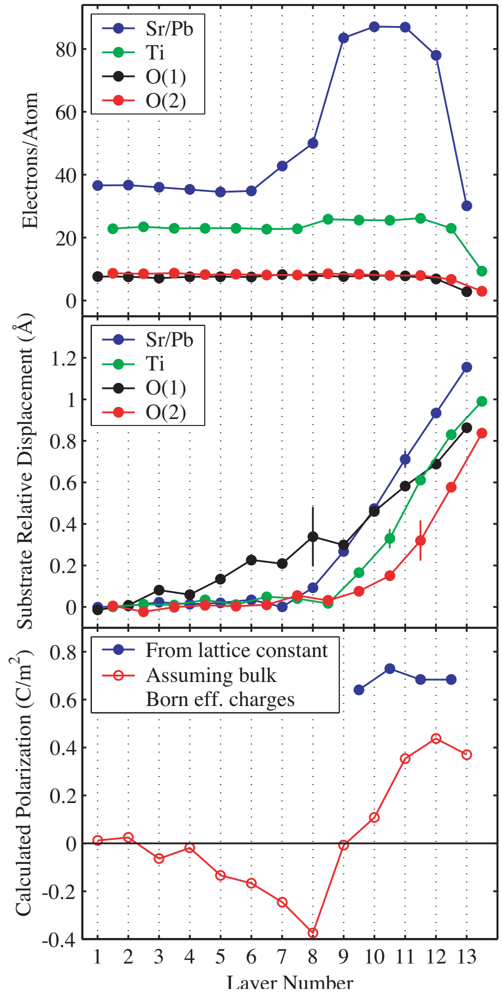


Fig. 6: Summary of COBRA-determined structure and derived polarization as a function of layer number. (a) Electron density at each atomic site. The nominal interface is between layers 8 and 9. (b) Atomic displacements relative to an ideal $SrTiO_3$ lattice. (c) Calculated polarization distributions using two different methods.

The proposed dynamic measurement of ionic displacements should shed light directly on this problem. This study of switching fatigue was suggested by some real-time domain imaging studies performed by our collaborator at Sector 7 Paul Evans (U. Wisconsin) and recently published in Nature Materials.

The ultimate aim of our work on ferroelectric heterostructures is coherent control of ferroelectric structure using pulsed electric fields and also the electric fields generated by ultrafast lasers. For example, we would like to investigate whether it is possible to switch the polarization of the ferroelectric film using nonlinear optical interactions generated by a pulsed ultrafast laser. This will require implementing COBRA in an ultrafast time-resolved mode. Recent upgrades to the laser facilities at Sector 7 will be beneficial in performing these challenging experiments. We will be attempting some of these time-resolved measurements in the next phase of the proposed research program.

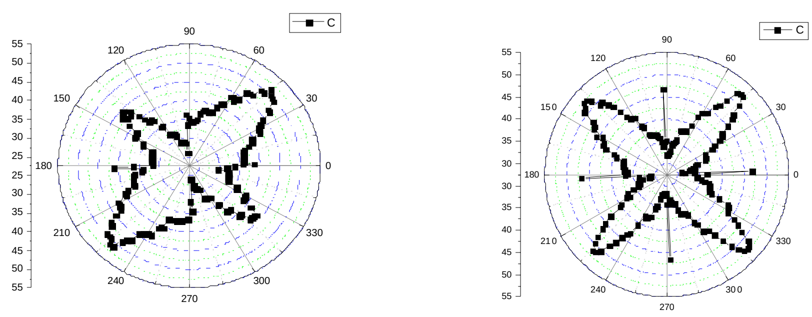
2.4 Studies of Ferromagnetic Heterostructures

Correlated structural and magnetization reversal studies on epitaxial Ni films grown with MBE and with sputtering

Graduate Students: Zhengdong Zhang[1], C. Cionca*[5]
Rosa A. Lukaszew[1]; A. Zambano[2] , M. Yeadon[3]; D. Walko[4]; E. Dufresne[4], R. Clarke[5]
*Supported by this grant
[1]Physics Department, University of Toledo, Toledo, OH
[2] Michigan State University, Lansing, MI
[3]National University of Singapore, Singapore
[4]Advanced Photon Source, ANL, IL
[5]University of Michigan, Ann Arbor, MI

Metal-ceramic interfaces are important in applications as diverse as catalysis, magnetic storage media and electrodes in spin-dependent tunneling junctions. It is important to understand how the crystallography and microstructure of metallic films deposited onto ceramic substrates depend on growth and/or annealing conditions so that their physical properties (e.g. magnetic, electronic, etc.) can be tailored for specific applications. To this end, we have studied the epitaxial growth and annealing of (001) and (111) Ni and FeN films grown on MgO substrates using different deposition techniques such as MBE (molecular beam epitaxy) and magnetron sputtering.

The evolution of the surface has been studied using correlated *in-situ* RHEED (reflection high-energy-electron diffraction), STM (scanning tunneling microscopy), TEM (transmission electron microscopy) and high resolution x-ray diffraction measurements.



(a) ***(b)***

Fig. 7: **Polar plot of coercive field determined with longitudinal MOKE for: *(a)* 30nm (001)Ni film MBE grown on MgO. *(b)* 30nm (001) Ni films sputtered on MgO. Note that only the 4-fold symmetry is evident in this sample.**

We have completed studies on the magnetic properties of these films, particularly the azimuthal dependence of the magnetization reversal (Fig. 7) as determined by longitudinal MOKE (Magneto-Optic Kerr Effect), and part this research project is to correlate these findings with the structural characterization obtained with *ex-situ* STM, TEM as well as x-ray diffraction studies performed at Sector 7 of the Advanced Photon Source.

From the azimuthal dependence of coercive field and the reciprocal space maps obtained with high resolution x-ray diffraction, we found that Ni films deposited under identical conditions on MgO with MBE and with sputtering are epitaxial, and have the same average coercivity due to the film's structure. Only the film grown with MBE shows additional uniaxial anisotropy, which we believe is due to a particular surface morphology characteristic of MBE growth.

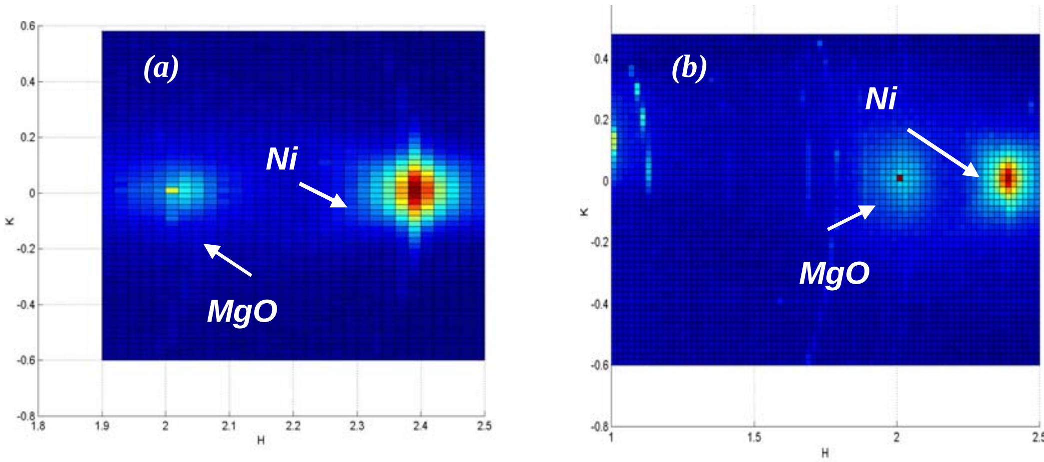


Fig. 8: Reciprocal space maps obtained with high-resolution X-ray diffraction, showing epitaxial cube-on-cube growth for: (a) 30nm Ni film MBE grown; (b) 30nm Ni films sputtered on (001) MgO.

From reciprocal space maps similar to those shown in Fig. 8 we are able to determine the lattice mismatch and consequent strain in the thin film samples, as well as the mosaicity. We find excellent correlation with the magnetoelastic contribution to the in-plane magnetic anisotropy shown in Fig. 7 for the MBE and sputtered samples, however we have not yet been able to resolve the structural origin of the uniaxial contribution evident in Fig. 7(a). High resolution cross-sectional TEM measurements have recently revealed the existence of a new phase that forms at this interface. We believe that this new interface structure, which is lower symmetry than cubic bulk Ni, is responsible for the uniaxial anisotropy. There is preliminary evidence, published by us in the International Journal of Nanoscience, that the new phase in question is an interface oxide.

Further experiments are in progress using glancing angle diffraction geometry and the COBRA technique to probe the interface region in more detail.

2.5. Instrumentation Development at 7-ID

Some of the materials research performed under the current funding has contributed significantly to improving the experimental capabilities of the beam line. Here we give one example, relating to the development of refractive focusing optics based on parabolic x-ray lenses made from lithium. These lenses have been used for some of the ultrafast x-ray diffraction experiments where paraxial focusing devices are advantageous on account of preserving the coherence and time structure of the probe x-ray beam.

2.5.1. Parabolic lithium refractive x-ray optics

Graduate student: Pedro Encarnacion,*
*Undergraduate Researcher: William Schlotter**

*Supported by this grant

Excellent x-ray optics for photons at around 10 keV can be expected with lithium metal. One of the best compound refractive lens designs is now produced routinely in aluminum, and more recently has been demonstrated using beryllium. Here, we report a similar refractive lens made from lithium. At 10.87 keV, this lens has a .2 m focal length, more than 90% peak transmission, and an average transmission of 49%. The lens shows a very useful gain of up to 40. The full widths at half maximum (FWHM) of the focus are blurred by roughly 20 μm, resulting in a horizontal and vertical FWHM of 33 and 17 μm for an image distance of 2.13 m. The lens produces speckle on the x-ray beam, which is likely due to the inhomogeneities of the lens surface: Coherent x-ray scattering is useful in understanding imperfections in x-ray optics, such as mirrors and lenses. Better molding techniques should result in improved performance and enable microbeam techniques with this type of Li lens.

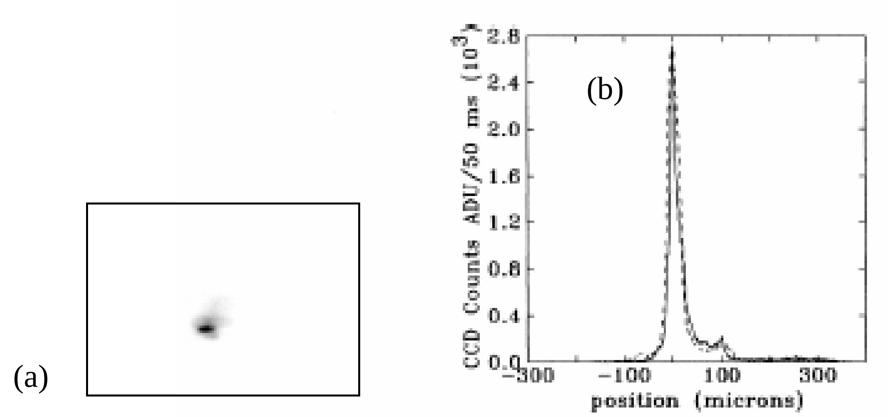


Fig. 9: (a) An inverted gray scale image of the focused x-ray beam in the image plane of the parabolic lithium lens; (b) cross sections through the focus, along *x* (dashed line) and *y* (solid line). The pixel size is 0.93 μm. The image horizontal and vertical FWHM is 33 and 17 μm, respectively.

The high quality of these lenses is further demonstrated in Fig. 10, comparing the focal spot of an undulator pink beam fundamental (~8 keV) with the higher harmonics which pass through the lens without significant convergence at this x-ray energy (> 24 keV).

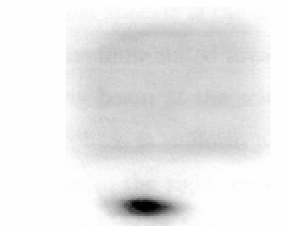


Fig. 10: Focused fundamental x rays below transmitted higher harmonic.

On the strength of these results, our industrial collaborator, Dr. Nino Pereira, was granted a Phase II SBIR grant to further develop Li-based x-ray optics. Access to beam time and the x-ray expertise at MHATT-XOR was instrumental in Ecopulse getting funding for this work enabling the company to commercialize this technology.

3. List of Publications Submitted or in Press

Refereed Journals

(titles in blue can be downloaded from the MHATT-XOR website: www.mhatt.aps.anl.gov/research/publications/papers.html

1. X-ray synchrotron studies of ultrafast crystalline dynamics, M.F. DeCamp, D.A. Reis, D.M. Fritz, P.H. Bucksbaum, E.M. Dufresne and R. Clarke, J. Synch. Rad., **12**, 177-192 (2005).

2. Direct Structure Determination in Ultrathin Ferroelectric Films by Analysis of Synchrotron X-ray Scattering Measurements , D. D. Fong, C. Cionca, Y. Yacoby, G.B. Stephenson, J.A. Eastman, P.H. Fuoss, S.K. Streiffer, C. Thompson, R. Clarke, R. Pindak, and E.A. Stern, Phys. Rev. B **71**, 144112 (2005).

3. Matrix-Seeded Growth of InGaAs:N Semiconductor Nanostructures Using Ion Beams", X. Weng, W. Ye, S.J. Clarke, and R.S. Goldman, V. Rotberg, A. Daniel and R. Clarke, J. Appl, Phys. 97, 064301 (2005).

4. COBRA Measurements of Interface Structure in GaSb-InAs Heterostructures, C. Cionca, C. Dorin, B. Perez, J. Mirecki Millunchick, D. Walko, and R. Clarke, Phys. Rev. B (submitted).

5. Parabolic lithium refractive optics for X-rays., N.R. Pereira, E. M. Dufresne, R. Clarke and D.A. Arms, Rev. Sci. Instrum. **75**, 37-41 (2004).

6. Correlated structural and magnetization reversal studies on epitaxial Ni films grown with molecular beam epitaxy and with sputtering, Z. Zhang, R.A. Lukaszew, C. Cionca, X. Pan, R. Clarke, A. Zambano, D. Walko, E. Dufresne, S. te Velthius, J. Vac. Sci. Technol. A 22(4), p1868-1872 (2004).

7. Surface morphology, structure and magnetic anisotropy in epitaxial Ni films, Rosa Alejandra Lukaszew, Zhengdong Zhang, David Pearson, Vladimir Stoica* and Roy Clarke, *J. of Alloys and Compounds*, **369**, 213-216 (2004).

8. Surface Morphology and Magnetization Reversal, R. A. Lukaszew. Z. Zhang, C. Cionca*, V. Stoica* and R. Clarke*, *J. Vac. Sci. Technol.* A **21**, 1524 (2003)

9. Supersonic strain front driven by a dense electron-hole plasma, M.F. DeCamp, D.A Reis, A. Cavalieri, P.H. Bucksbaum, R. Clarke, R. Merlin, E.M. Dufresne, D.A. Arms, A.M. Lindenberg, A.G. MacPhee, Z. Chang, B. Lings, J.S. Wark, and S. Fahy, Phys. Rev. Lett. 91, 165502-1 to 165502-4 (2003).

10. Effects of Three-Dimensional Forces in Topographical Imaging of Atoms with an Atom Force Microscope, W. Wang, S.J. Hu, and R. Clarke, Phys. Rev. B **68**, 245401 (2003).

11. Refractive Optics Using Li Metal, D.A. Arms, N.R. Pereira, E.M. Dufresne, R. Clarke, S.B. Dierker, and D. Foster, Rev. Sci. Instrum. **73**, 1492-1494 (2002).

12. "Coherent Control of Pulsed X-ray Beams", M. F. DeCamp, D. A. Reis, P. H. Bucksbaum, B. Adams, J. M. Caraher, R. Clarke, C. W. Conover, E. M. Dufresne, R. Merlin, V. Stoica, and J. Wahlstrand, Nature **413**, 825 (2001); *Search and Discovery*, Physics Today February, p. 16 (2002).

13. A Study of Concentration Fluctuations in the Binary Mixture Hexane-Nitrobenzene with X-ray Photon Correlation Spectroscopy , E.M. Dufresne, T. Nurushev, R. Clarke, S.B. Dierker
Physical Review E 65, 065107 (2002).

14. Direct determination of epitaxial interface structure in Gd_2O_3 passivation of GaAs Yizhak Yacoby, Mukhles Sowwan, Edward Stern, Julie Cross, Dale Brewe, Ron Pindak, John Pitney, Eric M. Dufresne and Roy Clarke, Nature Materials **1**, 99-101 (2002).

15. Direct atomic structure of epitaxially grown films: Gd_2O_3 on GaAs (100) M. Sowwan, Y. Yacoby, J. Pitney, R. MacHarrie, M. Hong, J. Cross, D.A. Walko, R. Clarke, E.A. Stern, Phys. Rev. B **66**, 205311 (2002).

16. Evolution of Structural and Optical Properties of Ion-beam Synthesized GaAsN Nanostructures, X. Weng, S. J. Clarke, W. Ye, S. Kumar, R. S. Goldman, A. Daniel, R. Clarke, J. Holt, J. Sipowska, A. Francis and V. Rotberg, J. Appl. Phys. **92**, 4012 (2002).

Encylopedia Article

Third-Generation Synchrotron Sources, R. Clarke, Encyclopedia of Modern Optics, Eds. B.D. Guenther, D.G. Steel, L. Bayvel, Elsevier, Amsterdam (2005).

Refereed Conference Proceedings Publications

1. Imaging and manipulating heterostructure interfaces (Keynote Address), Roy Clarke, Codrin Cionca, Catalina Dorin, Benny Perez Rodriguez, Joanna Mirecki-Millunchick, Don A. Walko, Yizhak Yacoby To appear in the Proceedings of the SPIE International Conference on Microstructures for the Millenium, Symposium on Nanotechnology, Seville, Spain, May 2005.

2. Surface nano-patterning effects, structure and magnetic properties of epitaxial Ni films", R. A. Lukaszew, Zhengdong Zhang, Dave Pearson, Vladimir Stoica, Roy Clarke and Mark Yeadon, accepted for publication in *International Journal in Nanoscience* (2005).

3. Experience with a Fluorescence-based Beam Position Monitor the Advanced Photon Source, E. M. Dufresne, J.A. Guzman, S.B. Dierker, R. Clarke, D.A. Arms, and D.A. Walko Proceedings of the International Synchrotron Radiation Instrumentation Conference SRI2003 in San Francisco August 2003, AIP Conference Proceedings vol 705, Melville New York, p679-682, April 6 (2004).

4. An Imaging System for Focusing Tests of Li Multi-Prism X-ray Refractive Lenses E. M. Dufresne, D.A. Arms, N.R. Pereira, P. Ilinski, and R. Clarke Proceedings of the International Synchrotron Radiation Instrumentation Conference SRI2003 in San Francisco August 2003, AIP Conference Proceedings vol. 705, Melville New York, p780-783, April 6 (2004).

5. Diffractometer-control software for Bragg-rod measurements, Y. Yacoby, D.A. Walko, M. Sowwan, R. Clarke, R. Pindak, R. MacHarrie, J.A. Pitney, E.A. Stern, and D. Brewe, Proceedings of the International Synchrotron Radiation Instrumentation Conference SRI2003 in San Francisco August 2003, AIP Conference Proceedings, 705, Melville New York, p1221-1224, April 6 (2004).

6. Large aperture x-ray refractive lens from lithium., N.R. Pereira, E. M. Dufresne, R. Clarke, D.A. Arms, Proceedings of the SPIE conference in Denver, CO Aug 2-6, 2004 Vol. 5539 p174-184 (2004).

7. Direct determination of the stacking order in Gd_2O_3 Epi-layers on GaAs Y. Yacoby, M. Sowwan, R. Pindak, J. Cross, D. Walko, E. Stern, J. Pitney, R. Macharrie, M. Hong, R. Clarke, Mat. Res. Soc. Symp. Proc. **751,** pZ4.5.1 to Z4.5.6 (2003).

8. Direct determination of epitaxial film and interface structure: Gd_2O_3 on GaAs (100), Y. Yacoby, M. Sowwan, E. Stern, J. Cross, D. Brewe, R. Pindak, J. Pitney, E.B. Dufresne, and R. Clarke, In the Proceedings of the Seventh International Conference on Surface X-ray and Neutron Scattering, September 23-27 2002 in

Lake Tahoe CA, USA, Ed. J.F. Ankner, S. Brennan, J. Kortright, Physica B **336** 39-45 (2003).

9. Design and performance of a stable first crystal mount for a cryogenically cooled Si monochromator at the APS., E.M. Dufresne, D.A. Arms, S.B. Dierker, R. Clarke, Y. Yacoby, J. Pitney, B. MacHarrie, R. Pindak, Rev. Sci. Instrum. **73**, 1511-1513 (2002).

10. Picosecond time-resolved x-ray diffraction probe of coherent lattice dynamics (abstract) (invited), D.A Reis, M.F. DeCamp, P.H. Bucksbaum, R. Clarke, E. Dufresne, and R. Merlin, Rev. Sci. Instrum. 73, 1361 (2002).

11. The Dynamics of Magnetorheological Elastomers Studied by Synchrotron Speckle Analysis, W.F. Schlotter, C. Cionca, S.S. Paruchuri, J.B. Cunningham, E. Dufresne, S.B. Dierker, D. Arms, R. Clarke, J.M. Ginder, and M.E. Nichols, in the Proceedings of the Eighth International Conference on Electrorheological Fluids and Magnetorheological Suspensions in Nice, France, 2001, published in the International Journal of Modern Physics B, Vol. 16, Nos 17&18 (2002) p2426-2432.

Conference Presentations

1. Imaging and manipulating heterostructure interfaces (KEYNOTE ADDRESS), Roy Clarke, Codrin Cionca, Catalina Dorin, Benny Perez Rodriguez, Joanna Mirecki-Millunchick, Don A. Walko, Yizhak Yacoby To appear in the Proceedings of the SPIE International Conference on Microstructures for the Millenium, Symposium on Nanotechnology, Seville, Spain, May 2005.

2. "Magnetic Nanostructures", R. Clarke*, (INVITED TALK) International Conference on Advanced Materials and Nanostructures, Wellington, New Zealand, February 2003

3. "Applications of Coherent X-ray Scattering", R. Clarke*, (INVITED TALK) International Workshop on
Coherent and Inelastic X-ray scattering, University of British Columbia, Vancouver
Canada, April 2003.

4. "Imaging Epitaxial Films and Interfaces with Sub-Angstrom Resolution: Coherent Bragg Rod Analysis (COBRA)", (INVITED TALK) Y. Yacoby, M. Sowwan, J. Pitney, R. MacHarrie, M. Hong, R. Clarke*, C. Cionca*, E. Stern, R. Pindak, D. Fong, B. Stephenson, J. Cross, D. Brewe, D. Walko, E. Dufresne*, X-ray Gordon Conference, Newport, RI, USA, July, 2003.

5. "Time resolved X-ray Scattering", (INVITED TALK) R. Clarke, XII International Conference on Advanced Materials, Cancun, Mexico, August 2003.

6. "COBRA: a new technique for Direct Structure Determination at Interfaces", INVITED TALK) R. Clarke, Annual Meeting of the American Vacuum Society, November 2003.

7. "Epitaxial Ferroelectric Heterostructures" R. Clarke, (INVITED TALK) International Materials Research Society, Singapore, December, 2003.

4. Ph.D. Theses Based on Work Supported by this Grant

Imaging Interfaces in Epitaxial Heterostructures.
C. Cionca
Thesis Advisor: Roy Clarke
Department of Physics, University of Michigan, January 2005

Coherent X-ray Scattering Measurements of Magnetorheological Composites
Jevne Micheau Cunningham
Thesis Advisor: Roy Clarke
Applied Physics Program, University of Michigan, December 2004

Electronic and Structural Properties of Boron Nitride Thin Films
Abishai Daniel
Thesis Advisor: Roy Clarke
Applied Physics Program, University of Michigan, July 2005

Investigation of solid-solid interface structure using a novel X-ray diffraction method.
M. Sowwan
Thesis Advisor: Prof. Y. Yacoby
May 2003
Department of Physics, Hebrew University, Jerusalem, Israel